Study on Chemical Variation of Interface Between Gd-doped UO_2 and Zr Through Annealing With Various Temperatures

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1. Introduction

The interaction between UO_2 and Zr has been studied to comprehend the interfacial reaction between the nuclear fuel and the cladding [1-3]. Especially, their interaction at high temperature has been considered in order to understand the situation for the loss-of-coolant accident (LOCA). Various fission products generated from the irradiation of the UO_2 fuel and could affect the physical and chemical properties of UO_2 and Zr. Therefore, the influence of temperature and fission product on the interfacial reaction between UO_2 and Zr should be important.

Among the various fission product, Gd has often been chosen as a dopant in simulated spent nuclear fuel and easily forms solid solutions with UO_2 [4-6]. It is also important element which is used as a burnable absorber. In this study, we selected Gd as a representative fission product.

Herein, we demonstrate the chemical variations at the interface between Gd-doped UO_2 and Zr before and after annealing at various temperatures using X-ray diffraction (XRD), scanning electron microscopy (SEM), and Raman spectroscopy.

2. Experimental

UO_2 and Gd_2O_3 powders were mixed to fabricate Gd-doped UO_2 ($U_{1-y}Gd_yO_2$) pellets with various compositions (0, 2, 6, and 10mol% Gd). Mixed powders were compacted into a pellet form (6.35 mm diameter). Compacted pellets were sintered in an alumina tube at 1700°C for 18 hours under H_2 atmosphere. Disc type Zr sample having a diameter of 6.35 mm was produced by cutting a Zr rod (Sigma Aldrich Korea Ltd). The Gd-doped UO_2 pellet was placed on a Zr sample so that they physically contacted each other. Annealing was performed for 10 minutes after reaching intended temperatures of 300, 700, and 1200°C in HTK-2000N chamber (Maintain pressure below 5×10^{-6} torr) respectively. After annealing, the interfacial surfaces of the Gd-doped UO_2 pellet and a Zr sample were analyzed. The SEM experiments were fulfilled using a JEOL JSM-6610LV with an Oxford Instruments EDS. Raman spectroscopy was performed using ANDOR Shamrock SR500i Raman spectrometer (He-Ne laser with a wavelength of 632.8 nm). XRD data were measured by Bruker-AXS D8 Advance system (Cu K_α radiation) in the 2θ range of 20 to 120° with a scanning step of 0.02° for 0.1 s. The lattice parameter was calculated using the Bruker TOPAS program.

3. Results

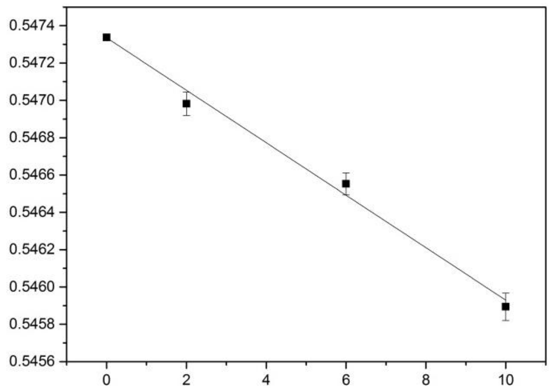

Fig. 1. The calculated lattice parameter of $U_{1-y}Gd_yO_2$ (y = 0, 2, 6, and 10) at Room temperature.

The calculated lattice parameters of the Gd-doped UO_2 at room temperature are shown in Fig.1. As the Gd concentration in the sample increased, the lattice parameter decreased. The similar features are observed at 300, 700, and 1200°C. There is no significant change in XRD data of Zr samples before and after annealing.

The Raman spectra of Gd-doped UO_2 after annealing at various temperatures show similar results each other. There are two main peaks at $445cm^{-1}$ and $1150cm^{-1}$. Those peaks are the fingerprint of the UO_2 fluorite structure. The intensities of peaks at 445 and 1150 cm^{-1} decreased with increasing Gd doping level. However, it is hard to find significant difference in Raman spectra between before and after annealing. It is expected that Gd doping could inhibit the reaction between UO_2 and Zr or the temperature may not be high enough to cause an interfacial reaction.

4. Conclusions

We simulated the interfacial reaction between Gd-doped UO_2 and Zr at various temperatures. The interface was analyzed using XRD and Raman analysis, but no specific changes were observed before and after annealing. Gd doping could lower the reactivity of the surface of UO_2. The higher temperature may be require to occur the the interfacial reaction between Gd-doped UO_2 and Zr. We will discuss the effect of Gd doping on the interfacial reaction in detail.

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